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W6-40517

DEVELOPMENT OF A WAVELENGTH-INDEPENDENT
RADIATION MONITORING FILM

FINAL REPORT

August 1, 1953

Work conducted by ANSCO, a Division of General
Aniline and Film Corporation, Binghamton, New
York, under Subcontract 31-109-38-139 with the
Argonne National Laboratory.

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Operated by The University of Chicago
under
Contract W-31-109-38-193

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4 DEVELOPMENT OF A WAVELENGTH-INDEPENDENT RADIATION MONITORING FILM

SUMMARY

[The purpose of this project was to develop a photographic film, or combination of films, sensitive in the range 0.1 roentgen to 30.0 roentgens as measured in air, the sensitivity expressed in reciprocal roentgens to be as nearly independent of wavelength as possible in the energy range of approximately 20 kiloelectron volts to 1 million electron volts. During the course of this work 15 written reports on progress were made to the Argonne Laboratory. This is the final report.

The first efforts to reach the desired goal consisted of modification of available commercial emulsions to reduce the dependence of sensitivity on wavelength. Although considerable progress was made using this approach, it was later abandoned in favor of a system involving a combination of a film and a pair of fluorescent intensifying screens.

The film and screens were designed so that the response of the combination is very nearly wavelength independent in the desired range. In addition to having the proper balance between sensitivity to the fluorescent light from the screens and sensitivity to direct x-radiation, it is also important that the film have a minimum of low-intensity reciprocity law failure to minimize the variation in sensitivity with exposure time in the range 1 sec to 40 hr. The film designed under the contract meets the requirements when processed in a special developer consisting of Ansco Liquadol x-ray developer and a developer retarder. The intensifying screens are pressure-molded from a mixture of p-terphenyl and Lucite. The fluorescence efficiency of the screens is greatest for high energy radiation - to which the film alone is least sensitive - and least for low-energy radiation - to which the film alone is most sensitive. This complementary relation between the fluorescence efficiency of the screens and the film sensitivity is a major factor in producing wavelength independence.

The film-screen combination fills the major requirements for radiation monitoring in the laboratory, in manufacturing areas and in the field. In the effective energy range from 20 Kev to 1 Mev, for doses from 0.14 to 1.2 roentgens, the maximum error in measurement is approximately $\pm 30\%$. The range of exposures which can be measured is approximately 0.02 to 10 roentgens. The upper limit can be extended to 30 roentgens by a slight increase in emulsion thickness.]

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I. INTRODUCTION

The response of a photographic emulsion to x-rays and gamma-rays is primarily a function of the mass absorption of the silver halide. Since the mass absorption coefficient of silver bromide varies from about 50 at a wavelength of 0.48 A (25 Kev) to 0.05 at 0.01 A (1.2 Mev), one might expect the sensitivity of a photographic film, expressed in terms of equal incident energy, to vary in a similar manner - roughly by a factor of 1000. In reality the variation is less pronounced. Only the truly absorbed energy, that is, the energy of the photo- and recoil electrons transmitted to the emulsion grains, contributes to latent image formation. The amount of truly absorbed energy for equal mass absorption varies appreciably with wavelength, being generally higher for high-energy photons. Furthermore, the ionization per grain is a function of the electron energy, causing overexposure of the individual grains by long wavelength x-rays and somewhat better utilization of this energy towards formation of developable image in the short wavelength range. The sensitivity variation of a non-screen type x-ray film is therefore only about 250:1 in the indicated range.

In photographic dosimetry it is customary to express film sensitivity in terms of exposure in roentgens. The roentgen unit of dose is based on the absorption of radiation per unit mass of air. In the range in which we are primarily interested, the variation in the mass absorption of air is only about 1/10 the variation for silver halide. Therefore, if one wants to express the sensitivity of a photographic film in reciprocal roentgens, that is, the reciprocal of the exposure in roentgens needed to produce a given photographic effect, one should expect a sensitivity variation of only about 25:1 between 0.48 and 0.01 A.

This is actually the case, as shown in Figure 1, which demonstrates the spectral sensitivity of a commercial double-coated non-screen x-ray film for exposure to filtered x-ray and gamma-ray radiation. All ordinary photographic films respond in a similar manner, although the absolute sensitivity level may vary greatly, depending on the inherent sensitivity of the grains, the grain size and the silver halide concentration per unit area.

It is evident that the use of such films for personnel monitoring without further precautions may lead to errors of great magnitude (up to 2500%). This shortcoming of the photographic method was of course recognized during the early stages of its use, and various ways and means were worked out to reduce the errors. One monitoring badge developed during the war uses a silver or cadmium filter in conjunction with two pieces of x-ray films of different sensitivity.^{1,2} The filter reduces the wavelength dependence by partial elimination of the soft part of the radiation; two films are used in order to obtain sufficient latitude. Another badge employs a film of somewhat flatter gradation and therefore greater latitude; in this case, too, a cadmium filter is part of the badge.^{3,4} In a third dosimeter the use

Fig. 1. Spectral sensitivity of typical non-screen industrial x-ray film.

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of several metal filters covering different areas of the film is recommended, permitting evaluation of the energy and of the dosage of the exposing radiation.⁵ If it is necessary to monitor very high dosage ranges, a combination of several films and filters might be of considerable value.⁶ Numerous other photographic dosimeters have been described, the majority of which deal with applications to specific problems such as the monitoring of beta-radiation or of high-intensity gamma-radiation and others.^{7,8,9}

The use of metal filters goes a long way towards achieving wavelength-independent response, as can be seen readily in the lower curve of Figure 1 which gives the response of the non-screen type x-ray film exposed with a cadmium filter of 1 mm thickness. Whereas without filtration the spectral sensitivity varies by a factor of more than 25, the cadmium reduces this variation to about 5 for effective wavelengths shorter than 0.25 Å. At longer wavelengths the filtration by cadmium is excessive, leading to a very rapid drop in sensitivity; this shortcoming is only partially compensated by the insertion of a window in the filter, permitting free incidence of the unmodulated radiation.

Realizing the advantages and disadvantages of the photographic method, it was the objective of the present project to develop an improved photographic dosimeter of essentially wavelength-independent characteristics in the 20-Kev to 2-Mev energy range and of sufficient sensitivity and latitude to permit routine personnel monitoring. The accumulated dosage was to have a direct relation to a single density reading.

The exploratory approach taken during the first phase of work under this project consisted in a study of various types of emulsions and their modifications. The wavelength dependence of commercial emulsions was determined, and efforts were made toward their improvement by changing total thickness and silver concentration, as well as the silver halide/gelatin ratio. In particular, work was done to explore the possibility of obtaining a system in which the energy absorption would take place predominantly in gelatin, the silver salt playing primarily the role of the recording medium. In another approach, efforts were made to increase the sensitivity of the silver halide grains to high-energy electrons, in order to reduce wavelength dependence by more nearly complete utilization of all the grains exposed by high-energy particles. During this phase a number of inorganic and organic sensitizers were investigated. Parallel with these experiments, the effects of overcoating an ordinary x-ray emulsion with photographically insensitive silver halides and with other metal halides were studied in an effort to supply an absorber of soft x-rays and an intensifier of hard x-rays. The addition of quantities of alkali halides with traces of thallium to the finished emulsion was also tried, in the hope that the incorporation of a primitive scintillation material would help.

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Almost every one of the above-mentioned steps led to noticeable improvements. However, keeping in mind the necessity for a simple and easily workable solution, the progress achieved was not substantial enough. By combining several of the favorable effects one could expect, at best, to achieve a sensitivity ratio of 8:1, perhaps 6:1, at the expense of greatly complicating emulsion manufacture.

Subsequently a determined effort was made to utilize organic scintillation crystals as fluorescent intensifying materials. At the time of the initiation of this phase of the project, relatively little quantitative information of the wavelength dependence of these materials was known. However, it was soon learned, simply by exposing small quantities of such materials in contact with a blue-sensitive film, that many of these compounds exhibited much stronger fluorescence when exposed to the gamma-rays of radium than to soft x-rays. Various ways of combining the most efficient scintillation materials with the photographic emulsion were tried until, finally, a pressure-molded terphenyl-Lucite screen was evolved as a practical and efficient intensifier in the desired wavelength range.

Simultaneously a suitable emulsion was modified and adapted to the screen for appropriate speed and wavelength dependence by adjusting the silver/gelatin ratio and thickness. Considerable difficulty arose, however, in efforts to overcome reciprocity law failure, a characteristic common to all ordinary photographic emulsions. Whereas this failure does not play a significant role in direct x-ray exposures, it is very pronounced for exposures to light. Since, in the proposed badge most of the exposure at short wavelengths is due to light emitted by the screens, the effects of reciprocity law failure had to be eliminated. In efforts to do this it was particularly difficult to fulfill the essential requirement of emulsion stability with age. A suitable emulsion was finally designed by modifying a negative emulsion of moderate speed, particularly by supplying more thermal energy during the manufacturing stages and by enhancing chemical sensitization. Organic development retarders, which were added to a commercial developer for the processing of this emulsion, improved reciprocity characteristics further and keep the fog level low.

The final badge which resulted from this work is believed to fill all the major requirements which are essential for unrestricted use in the laboratory, in manufacturing areas, and in the field.

II. EMULSION MODIFICATIONS TO INCREASE GAMMA-RAY SENSITIVITY

A. Physical Basis

This chapter deals with efforts to decrease the wavelength dependence of photographic film by increasing the intrinsic sensitivity of the silver halide grains to high-energy photo- and recoil electrons. As discussed in the previous chapter, it is primarily the true absorption of the silver halide (that part of the absorbed energy which is transmitted to the silver halide grains) which determines the wavelength dependence. Another important factor is the efficiency with which the truly absorbed energy is used for latent image formation. The maximum efficiency would be obtained if the emulsion were sufficiently sensitive so that every grain hit by a photo- or recoil electron were rendered developable. In analyzing this further, one must consider the following pertinent basic facts:

1. The range of the secondary electrons in the emulsion layer increases rapidly with increasing energy, and exceeds the thickness of ordinary emulsions for energies well within the energy band under investigation. This is evident from Figure 2 which shows the range of electrons in pure silver bromide. (The thickness of a single layer of fast commercial x-ray emulsions is of the order of 30 microns. If the gelatin is eliminated, the thickness of the silver halide alone is about 4 microns.)

2. The energy loss of electrons per unit path-length is a function of the electron's energy. Low-energy electrons produce high specific ionization; for example, a 14-Kev electron can lose all of its kinetic energy in traversing a silver halide grain of about 1 micron diameter, thereby producing about 2400 ion pairs.¹⁰ In contrast, a 1-Mev electron loses energy at a rate close to the minimum given by the energy-loss curve (Figure 2), transmitting to the 1-micron grain only about 1 Kev which, in turn, produces about 170 ion pairs.

If one applies these latter observations to a fast commercial x-ray emulsion of 0.75-micron average grain diameter, one arrives at exposure extremes of about 12 Kev and 0.75 Kev, corresponding to the production of 2000 ion pairs in the first case and 130 in the second, assuming single direct hits only. In reality, because of the randomness of the hits, there is no lower limit for the ionization occurring per grain, a high percentage receiving exposures of much less than 0.75 Kev.

In a semi-quantitative study of two commercial Ansco x-ray emulsions, the threshold sensitivities of the average grains were found to be between 200 and 300 ion pairs.¹¹ Evidently, many grains receive much less than this amount and are consequently not rendered developable. The percentage of such grains must be much higher for exposures to x-rays of short wavelength

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Fig. 2. Rate of energy loss and range of electrons in pure AgBr.

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than to x-rays of long wavelength. Consequently, it seemed worthwhile to try to decrease the wavelength dependence of the film by increasing the intrinsic sensitivity of the grain to electrons.

B. Methods to Increase Intrinsic Sensitivity

In order to understand fully the steps taken in efforts to increase the sensitivity of the grain to electrons, it is necessary to review briefly a few basic photographic manufacturing procedures and the mechanisms of latent image formation and sensitization.

In conventional emulsion manufacture, silver halide grains are precipitated by adding a solution of silver nitrate to a potassium or ammonium bromide solution containing moderate quantities of gelatin. The rate of addition, the temperature of the solutions (approximate range 50-75 C), the duration of holding after addition, the quantities of gelatin present and the amount of excess solvent used (bromide or ammonia) determine the grain size distribution and, to a certain extent, the sensitivity of the emulsion. The holding period directly after precipitation, during which Ostwald* ripening occurs, is usually called first ripening or digestion. Subsequently the emulsion is chilled, washed to remove soluble salts, remelted and then subjected to a second heat treatment (40-60 C, up to 2 hr). During this heat treatment, called second- or after-ripening, the sensitivity to light, and to a lesser extent to x-rays, increases greatly. Chemical sensitization takes place during both phases of the treatment, the sensitization occurring in the second phase usually being the more important.

The latent image is formed in the course of an electronic and ionic conduction process, as originally proposed by Gurney and Mott¹² and later modified in specific details by Mitchell and others.¹³ According to these theories, absorption of light or ionizing radiation produces conduction electrons which are trapped at the sensitivity centers of the crystal. The resultant negative charge of the centers is subsequently neutralized by slowly moving interstitial silver ions, forming metallic silver. Simultaneously, the positive holes (bromine located in bromide ion lattice sites), which are highly mobile,^{14,15} must also be trapped to prevent their recombination with conduction electrons. The metallic silver represents the latent image; an aggregate of about 5 to 10 silver atoms suffices to initiate development.

It is the function of the chemical sensitizers to provide sensitivity centers which can act as trapping sites for the conduction electrons as well as for their "holes."

*C. E. Kenneth Mees in The Theory of the Photographic Process (Macmillan Company - New York - 1942), p. 21.

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The best-known sensitizing agent is sulfur, which is present in gelatin in the form of allyl thiourea or allyl isothiocyanate, according to Sheppard,¹⁶ or in the form of thiosulfates and polythionates, according to more recent work by Wood.¹⁷ Sulfur is believed to act as an electron trap when associated with a bromide ion vacancy and as a hole trap when present as a doubly-charged sulfide ion in a bromide ion lattice position.¹⁸

Almost equally important as a sensitizer is silver iodide, which is present in commercial silver bromide emulsions in concentrations of a few per cent, greatly enhancing the sensitivity to light. Iodide is believed to produce strains in the silver bromide, the resulting lattice imperfections providing efficient electron-trapping sites. However, there exists also a possibility that groups of iodide ions in the lattice act as hole traps, thus enhancing the sensitivity by minimizing recombination.¹⁹

A third very effective sensitizer is gold. Addition of small quantities of gold complex salts in the form of aqueous solutions of ammonium or alkali aurous thiocyanates during the second ripening produces greatly increased sensitivity to light and x-rays.²⁰ Gold in association with silver is believed to provide very effective trapping sites at the surface of the grains, thus reducing formation of the latent image in the interior where it is inaccessible for ordinary development. It may also have a development enhancing effect.

Other known sensitizers are lead and thallium salts. Mueller found that the addition of very small quantities of lead nitrate during emulsion precipitation increased x-ray sensitivity, at the same time decreasing the sensitivity to visible light.²¹ The presence of doubly-charged lead ions in the crystal lattice in small concentration increases the number of silver-ion vacancies, decreasing at the same time the number of interstitial silver ions and of (positive) bromide-ion vacancies. One can assume tentatively that the sensitization effect for x-rays (electrons) is due to more efficient trapping of positive holes by the negative silver-ion vacancies, whereas the loss in light sensitivity is probably related to a decrease in electron-trapping sites (bromide-ion vacancies) and interstitial silver ions. Even less is known about the mechanism of the thallium sensitization; since thallium acts as a sensitizer for light and x-ray exposures, one must assume that in this case electron trapping is greatly enhanced.

Until more recently little was known as to how x-ray sensitivity was affected by most of these sensitizers. Naturally, earlier studies dealt primarily with light sensitivity, and only few workers were concerned with x-ray sensitization. In fact, the reviewer of the photographic literature gained the impression, at least in the years up to 1950, that the sensitivity to x-rays is hardly affected by chemical sensitization. Even less was known about the possibility that such sensitization was wavelength dependent.

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We were therefore confronted with the problem of whether to study in a broad fundamental way the response of photographic systems to ionizing radiation or rather to try immediately to adapt existing systems to the contract requirements. As attractive as the first choice might have been, it was felt that the second course should be taken, in view of the predominantly practical aspects of the objective.

The work during the first phase of the contract primarily consisted, therefore, in attempts to increase the emulsion sensitivity towards x-rays of short wavelength by increased chemical sensitization.

1. After-ripening

The effect of after-ripening on x-ray and gamma-ray* speed was investigated. It is speculated that ripening consists essentially of an annealing of the crystals, causing partial removal of defects from the interior of the crystal by their migration to the surface where they aggregate and react with silver sulfide formed by the sulfur of the gelatin, or with other chemical sensitizers, forming effective surface trapping sites. The presence of a sufficient number of effective surface traps is more essential in exposures to gamma-rays than to soft x-rays, for reasons discussed before. These theories were confirmed by the experimental data which indicated that ripening decreases the 65-Kvp/gamma-ray speed ratio. The improvements thus obtained are limited by the increase in fog with extremely high ripening.

2. Iodide Sensitization

According to Mees,²² the action of silver iodide in silver bromo-iodide emulsions is complicated. Silver bromide crystallizes in the face-centered cubic system, whereas silver iodide can exist in two forms, one of them being the body-centered cubic system. Coprecipitation of iodide therefore causes strains in the silver bromide lattice and changes in the external crystal shape. These crystal deformations are probably excellent sites for electron and hole trapping. Little seems to be known about the effect of iodide on x-ray response. Experiments made with several emulsions indicated that the 65-Kvp/gamma-ray speed ratio could be influenced by variations of the iodide concentration, the optimum concentration depending on the type of emulsion used.

3. Gold Sensitization

The application of this type of sensitization to x-ray emulsions had been explored by F.W.H. Mueller and the writer prior to the present contract.²⁰ Further studies were made of the possible applications of gold

*The term "gamma-ray," as used in this report, refers to the gamma-rays of radium.

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sensitization to the present problem. The fact that gold sensitization was found to be more effective for fine-grain emulsions and gamma-rays indicates strongly that the gold effect is mainly a surface effect. The sensitization for gamma-rays and light increases in proportion to the ratio of surface area to volume, as grain size changes. Apparently, the gold increases the number and depth of the surface traps for electrons. Fully conclusive proof of this hypothesis has, however, not yet been provided.

Supporting evidence can be derived from studies of the latent image distribution in unsensitized and gold-sensitized grains exposed to white light, gamma-rays and x-rays, as described by Larson²³ and by the writer.²⁴ Similar surface and internal developments were carried out with several emulsions which had been sensitized with gold to different degrees. Invariably, gold sensitization resulted in a very strong increase in surface sensitivity and a simultaneous moderate decrease in internal speed.

It has also been proposed by Berg,²⁵ and others, that the effect of gold sensitization might consist predominantly of a catalytic enhancement of development. There is very little evidence in support of this latter hypothesis; in fact the degree of development (longer duration, higher temperature, addition of silver halide solvents, etc.) can increase the speed of an unsensitized emulsion to as much as half the speed of the fully gold-sensitized emulsion.

It is the wavelength dependence of the gold sensitization which supports most strongly the theory that both the number and depth of surface traps for electrons are increased. In the case of soft x-ray exposures the exposed grains are actually flooded with conduction electrons and, in spite of its great dispersion, sufficient latent image is formed, even in the unsensitized emulsion, to initiate development. (The main reason why the older phototechnical literature denies any influence of chemical sensitization on x-ray sensitivity is because all of the early experiments were conducted with low-energy radiation.) At increasing energies, the amount of ionization per average grain decreases, and the need for more efficient permanent trapping of electrons and silver ions at the surface, or close to the surface, increases, hence the greater efficiency of gold sensitization for radiation of shorter wavelength.

4. Thallium and Lead Sensitization

Increased x-ray sensitivity due to thallium and lead added to x-ray emulsions during the precipitation stages has been found by Mueller.²¹ Further studies of the nature of the lead sensitization are currently being made in the Ansco laboratories.²⁶ In connection with the present project the prime objective was to learn the wavelength dependence of these effects. Experiments with several emulsions yielded only very moderate improvements in the 65-Kvp/gamma-ray speed ratio. In view of this fact, more extensive work along this line did not appear justified.

5. Electron Sensitization with Organic Compounds

Another approach to increasing the x-ray sensitivity at short wavelengths can be derived from the results of nuclear track work relating to the recording of full-length tracks of high-energy electrons.

Commercially available nuclear track emulsions which are designed to record high-energy electrons have a relatively low reciprocal roentgen speed because of their very fine grain. The limited keeping quality and a wavelength-dependent speed regression with age make them unsuitable for monitoring work. However, in a recent publication, L. Jenny²⁷ disclosed an apparently new method for the sensitization of photographic emulsions to high-energy electrons. According to Jenny, certain organic dye molecules can be ionized by high-energy electrons with subsequent migration of the ejected electrons to the sensitivity centers of the silver halide, thus enhancing latent image formation. Dyes which sensitize in this manner must carry electron-donating groups such as NH_2 , NHR , NR_2 or OH , and they should preferably possess heterocyclic nuclei (with N and S). They should not carry NO_2 , $-\text{N}=\text{N}-$, and COOH groups, which are suspected of decreasing or destroying the sensitizing effect. In particular, Jenny described a new derivative of pyrazole* as an extremely effective sensitizer, possessing a heterocyclic nucleus and the obligatory NR_2 group.

A sample of this dye was obtained from Switzerland,²⁸ and other organic compounds available at Ansco were canvassed for suitability. Altogether, 26 different compounds were tested as additions to the emulsion before coating. At least two concentrations were used in each case, the amounts added being usually 60 and 300 mg per 10 kg emulsion, which are the quantities recommended by Jenny. They are of magnitudes similar to those employed in optical sensitization. The Ansco samples were selected from different groups:

1. Polynuclear cationic dyes, with and without NH or NR_2 groupings. Several of the dyes selected are known to be weak optical sensitizers.
2. Compounds containing the required dialkyl amino phenyl groupings attached to cyclic and heterocyclic nuclei.
3. Substituted amine stilbenes, so-called "blancophors" or "brighteners." All of these are absorbers of ultraviolet radiation and fluoresce strongly between 4000 and 4800 Å; several filled, at least partially, the requirements outlined in the foregoing.
4. Diphenylamine, described by Jenny as an electron sensitizer of moderate efficiency.

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On the whole, the results of these experiments were somewhat disappointing. Only very moderate speed increases for gamma-ray and x-ray exposures could be observed; they amounted, at best, to an improvement of the 65-Kvp/gamma-ray speed ratio by 15%. In all cases, the observed sensitizations were very close to the experimental error. Details are described in Appendix 3.

In spite of the generally discouraging results of this approach, caution must be used in evaluating this method of sensitization on the basis of these experiments. It was observed that all compounds which produced a moderate positive effect possessed structures as postulated by Jenny. On the other hand, almost all compounds tested which did not conform with the requirements produced desensitization.

Therefore, it appears possible that electron sensitization is harder to achieve with the relatively coarse-grain, highly ripened emulsion employed here than with the fine-grain, low-ripened emulsion used by Jenny. However, because of the small initial effects no further efforts were made to work with other emulsions; it seemed more profitable to concentrate at this time on other more promising approaches to the main goal.

6. The Effect of Silver Concentration and Silver/Gelatin Ratio

The x-ray mass absorption coefficients of gelatin and of human body tissue are known to be very similar. The thought occurred therefore to design a system in which the absorption of the incident radiation would be primarily a function of the gelatin, the silver halide taking second place as primary absorber, acting predominantly as a recording medium. In other words, the system should consist of large quantities of gelatin surrounding widely dispersed silver halide grains. A major contribution to the exposure of the silver halide would come from the photo- and recoil electrons originating in the gelatin. Such a system should undoubtedly have a better wavelength dependence than the conventional ones. In exposure to soft x-rays, a high percentage of the electrons originating in the gelatin would not have sufficient range to reach the silver halide grains; therefore, their energy would be dissipated without producing any latent image. This effect would be considerably less for hard x-rays, because of the greater range of the ejected electrons.

In reality, the situation is more complicated. The mass absorption coefficients of silver halide and gelatin in the soft x-ray range differ greatly, calling for a very low silver/gelatin ratio. This low ratio, in turn, may cause excessive energy loss by high-energy electrons in the gelatin surrounding the silver halide grains. From the manufacturing and processing point of view the maximum coating thickness of such a system is limited to about 100 microns. Nevertheless, an attempt was made to learn how far the 65-Kvp/gamma-ray speed ratio could be improved by decreasing silver concentration, by increasing total coating thickness and by a combination of both methods.

Summarizing the results of these experiments, the 65-Kvp/gamma-ray speed ratio was decreased by as much as 30% by decreasing the silver halide/gelatin ratio by a factor of 10 and increasing the coating thickness by a factor of 2. The improvement, though pronounced, falls short of the goal, partially because the ideal case involves a coating thickness which presents considerable difficulty in manufacture, as well as in the processing of the exposed film.

C. Effect of Extended Development

As stated in the foregoing, the amount of latent image produced per average grain by x-rays is considerably less for exposures to radiation of short wavelengths as compared to exposures at longer wavelengths. In the case of gamma-rays of radium, many grains receive exposure corresponding to the minimum rate of energy loss (Figure 2) but are not rendered developable under ordinary circumstances. Therefore, experiments were conducted to enhance development by changes of developer formulation, in particular by the addition of greater-than-standard quantities of silver halide solvents such as sodium sulfite, and also by the addition of small quantities of strong solvents such as hypo ($\text{Na}_2\text{S}_2\text{O}_3$). The resulting increases in gamma-ray speed were moderate. The most beneficial effect was obtained simply by extending the development time with the commercial Liquadol Developer to six minutes. Liquadol is a very potent, caustic developer, formulated specifically for the development of x-ray emulsions.

The following 65-Kvp/gamma-ray speed ratios are illustrative of the results obtained:

Development Time (min.)	2	3	4	5	6
Speed Ratio	18.5	16	15	14	13.5

D. Other Exploratory Experiments

Numerous experiments of exploratory nature were conducted to investigate specific effects involving physical intensification, that is, systems containing absorbing media for soft radiation or fluorescent substances in the emulsion. Many of these trials were successful insofar as moderate improvements in wavelength dependence were obtained. However, the gains achieved were not of sufficient magnitude to justify extensive experimentation.

E. Summary of Preceding Experiments

The experiments described above were directed mainly toward the design of a modified, but still conventional, emulsion. It appeared that by the proper combination of the modifications studied, an emulsion could be made with a 65-Kvp/gamma-ray speed ratio of about 6, and the absolute sensitivity and latitude requirements of the contract would be met. Although this represents a great improvement over conventional commercial emulsions, it was felt that the achievement was still too far from the goal. In the meantime, work on another approach had become very promising, and all efforts were directed toward the second phase of the subject.

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III. INTENSIFYING SCREEN-FILM COMBINATION

A. Scintillation Materials for Intensifying Screens

In the postwar years the organic scintillation materials originally explored by Kallmann²⁹ found wide applications in nuclear physics instrumentation. They provided very useful tools in the investigation of individual events because of their strong light emission in pulse form as a function of particle absorption. There is, of course, no reason why they should not be applied also in connection with integrating receivers such as photographic film. In fact, the closeness of their average atomic number to that of air or human body tissue should make them ideally suited for dosimetry if a proper combination with the receiver could be found. Further, it appeared, and soon was verified, that the fluorescent efficiency of many of these materials was lower for soft radiation than for hard radiation, thus counteracting the effects of the absorption features of the film.

After it had become evident during the first phase of this project that emulsion modifications alone would not lead to a fully satisfactory solution, the use of organic scintillation materials was taken into consideration. During December, 1949, the first naphthalene screen was made by adding naphthalene to a gelatin solution. The screen, containing approximately 200 mg naphthalene per cm^2 , was placed in contact with a blue-sensitive film and exposed to gamma-rays of radium and to 65-Kvp x-rays. In the case of gamma-rays the film density in the area behind the screen was strongly enhanced, due to the fluorescent radiation emitted by the screen; in the case of 65-Kvp x-rays, the density was actually decreased, due to the absorption of the primary radiation. This experiment demonstrated the usefulness of scintillation materials for this particular application.

Subsequently, a systematic search was conducted for suitable organic materials. Since the search was to be qualitative, a very simple test procedure was developed which consisted of loading a flat cellophane bag with 100 mg/cm^2 of powdered or crystalline material and exposing a sheet of Ansco High Speed X-ray screenfilm in contact with the flat bag. The intensification factors (ratio of photographic speed with intensifier to speed without intensifier at net D = 1.0) were determined for gamma-rays and, where promising, also for 65-Kvp x-rays. Since the blue-sensitive x-ray screen film would not record the emission of green and red light, panchromatic film was used in a few cases.

The fluorescence under ultraviolet excitation was also observed visually. No correlation was observed between ultraviolet and x-ray or gamma-ray excitation.

135 selected samples were screened in this manner. Without going into specific details, the results can be summarized as follows:

1. Study of cyclic hydrocarbons

a. Polyphenyls. Of the series of hydrocarbons represented by

		Intensification factor*
Diphenyl		2
p-terphenyl		7
p-quadriphenyl		trace

the peak of relative effectiveness was obtained by p-terphenyl.

b. Fused rings. In a similar fashion, the linear arrangement of the three fused benzene rings in purified anthracene is by far the most effective structure in this series:

		Intensification factor*
Naphthalene		3
Anthracene		7
Phenanthrene		trace
Pyrene		trace
Chrysene		1.4

*All intensification factors are given for radium gamma-ray exposures. The factors for 65-Kvp x-rays were close to 1 in all cases.

The desirable intensification characteristics of naphthalene are not maintained when the structure is enlarged to that of acenaphthylene or changed to indene.

	Intensification factor*
Acenaphthylene	trace
Acenaphthene	trace
Indene	trace
Fluorene	2
Fluoranthene	1.5

c. Phenyl groups joined by saturated and unsaturated aliphatic chains.

	Intensification factor*
Diphenyl Methane	trace
Triphenyl methane	trace
Dibenzyl	5

	21	Intensification factor*
Trans Stilbene		6
Tolan		2
Distyryl		trace

In all of these cases, the purities of the members of the series, particularly of the unsaturated ones, appear to be critical. All of these substances are very susceptible to having their fluorescence effectiveness "quenched" by the presence of traces of impurities. The compounds used in this study were all purified, but absolute purity can only be assumed and care should be taken in arriving at theoretical conclusions.

2. Study of heterocyclic compounds, substituted cyclic hydrocarbons and synthetic resins. A rather large number of compounds were evaluated to determine the effect of various substituents in or on a ring structure. Generally, the results were discouraging; only a very few compounds retained any of the desirable characteristics of the parent hydrocarbon. Of the various resins tested, only two exhibited a slight-to-moderate intensification; the rest were ineffective.

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	<u>Intensification factor</u>
1-4 diphenyl butadiene	2.7
Polystyrene	2
Polyvinyl carbazole	1.6

Among the whole group, anthracene and p-terphenyl showed by far the most promising characteristics. The high efficiency of anthracene had been described earlier in the literature. The usefulness of p-terphenyl was discovered independently in the Ansco Laboratories shortly before Hofstadter, Liebson and Elliot's letter appeared in the Physical Review.³⁰ Since these compounds were not very well suited for our applications in the form of large single macro crystals, a study of the characteristics of their micro crystals in combination with carrier materials ensued.

B. Screen Manufacture

From our work with powders and also from a concurrent study of several commercial monocrystals,³¹ it was learned that at least 100-150 mg/cm² of a high-quality scintillation material are needed to obtain the desirable gamma-ray intensification and a concurrent minimal fluorescence under soft x-ray excitation. Anthracene is commercially available in high purity³² as scintillation-grade anthracene. The purity of commercial p-terphenyl³³ was not good enough to yield high fluorescence efficiency. Consequently, a recrystallization-purification process was worked out and is described in Appendix 4.

In our first approach, intensifying screens were produced by dispersing the scintillation material in four different carriers: gelatin, polyvinyl alcohol, cellulose acetate and methyl methacrylate. The mixtures employing the three first-mentioned carriers were coated or cast on glass or highly polished stainless steel plates. Depending on thickness, type of carrier and concentration of the fluorescent compound, the resulting foils showed various degrees of flexibility, the cellulose acetate foils being the most brittle ones. The coating and casting procedures for thicknesses in excess of 200 microns were quite cumbersome; it was difficult to obtain smooth coatings of constant thickness. Very soon, it became quite clear that conventional coating and casting methods of the type practiced commercially at Ansco could not be applied successfully for this purpose.

Methyl methacrylate, the fourth carrier, had been used to produce screens by first coating on plates a mixture of the scintillator and the monomer of the carrier and subsequently polymerizing the carrier. This carrier proved to be the most promising one because of the higher efficiency of the mixture as shown in Table I.

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TABLE I

Relative Efficiency of Organic Intensifying Screens
as a Function of Carrier Material

	<u>Gelatin</u>	<u>Polyvinyl alcohol</u>	<u>Cellulose acetate</u>	<u>Polymethyl methacrylate</u>
p-terphenyl	1	1.1	1.20	1.50
Anthracene	1	1.1	1.05	1.20

It was also learned that the quantities of scintillation material needed per cm^2 were quite large. For example, the intensification factor of a 200-micron anthracene-polyvinyl alcohol foil containing about 12 mg/cm^2 scintillation material was not more than 2.3 for a low-intensity gamma-ray exposure (1 hr exposure time). Since gamma-ray intensification factors of 15 or more were needed, the screen thickness and amounts of scintillation material had to be increased. Thicker screens of good efficiency were obtained with methyl methacrylate by the polymerization method; however, this process is not suited for large-scale application. Bubble formation, separation and sedimentation difficulties were encountered. Finally, upon S. Popeck's³⁴ suggestion, a pressure molding procedure using mixtures of the scintillator and the polymer of the carrier was worked out. This method yielded mechanically rigid, optically- and photographically satisfactory screens. The molding procedure is described separately in Appendix 5.

Before arriving at the final formulation, it was necessary to determine the optimum concentration of the scintillator in the carrier material. In a series of experiments, the concentration of scintillation material was varied from 10 to 300 mg/cm^2 , and the thickness was varied from 200 microns to 8 mm. The weight ratio, scintillation material/carrier, was varied from 0.1 to 1.0. The results of these tests showed that for a given concentration of scintillator per unit area (mg/cm^2), the highest fluorescence efficiency was obtained with the lowest scintillator/carrier weight ratio. (Whether this is due to a physical or an optical effect could not be ascertained. The possibility exists that at low concentrations part of the incident energy absorbed in the carrier is transmitted to the scintillator where it is re-emitted as fluorescence radiation; on the other hand, at high concentrations optical absorption of the fluorescent radiation in the scintillator itself must play an important role in limiting the efficiency - in this case, one of the functions of the carrier would be to "pipe" the light from the crystals to the film.)

Since, from the point of view of cost, it is best to obtain the desired intensification with the lowest concentration of scintillation material per unit area, it would be desirable to make relatively thick and transparent screens.

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However, the thickness of the whole film badge was not to exceed the dimensions of the standard badges, including filter, which are conventional in A.E.C. installations. A maximum thickness of 2.2 mm per screen was permitted.

This requirement, and the need for the highest possible intensification, led to a study of the relationship between the concentration of scintillation material for constant screen thickness (2.2 mm) and the intensification factor. The resulting data are shown in Figure 3 for molded terphenyl-methyl methacrylate ("Lucite") screens. The film used for these tests was coated with Ansco M-Type emulsion.*

Figure 3 shows that the light emission increases gradually with increasing p-terphenyl concentration to a maximum for 100 % terphenyl, represented by a slab of highly compressed terphenyl crystallites. In practice, a lower concentration has to be used for reasons of mechanical rigidity.

The screens which were finally selected - after considerable testing with the special emulsion coatings to be described later - contain 147 mg/cm² of p-terphenyl and 74 mg/cm² Lucite (polymerized methyl methacrylate), sandwiched between two identical polymerized Lucite layers which together are equivalent to 32 mg/cm².** Additional physical characteristics, including spectral transmission data, are given in Appendix 6.

The intensification characteristics of the screen as a function of the effective wavelength are plotted in Figure 4.† These data apply for the use of two screens, one on each side, in contact with the double-coated M-Type film for a low-intensity exposure yielding net D = 0.4, in one hour. The film was processed 3 min in a special developer (Liquadol-PMT) which is also described in a later paragraph.

*Available for restricted distribution by permission of Argonne National Laboratory.

**As noted in a preliminary report before the American Physical Society (H. Hoerlin and F. Kaszuba, Phys. Rev. 88, 174(A), 1952), the first screens made resulted in a dip in the film-screen response curve in the wavelength range from 0.06 Å to 0.14 Å. This deficiency was removed by the addition of 4 mg/cm² PbCl₂, after the suggestion of Drs. J. E. Rose and L. D. Marinelli of the Argonne National Laboratory. Later, as a result of improvements in the screen manufacturing technique and adjustment of the film emulsion thickness, the need for the PbCl₂ was eliminated.

†The intensification factor is defined as the ratio of the speed (in reciprocal roentgens) of the film with intensification to its speed without intensification.

Fig. 3. Gamma-ray intensification factors as a function of terphenyl concentration in terphenyl-Lucite screens.
Exposures made to M-type emulsion; exposure rate 0.14 r/hr.

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Fig. 4. Intensification factor for terphenyl screens and double-coated M-Type film.

Exposure time - 1 hr. Processed 3 minutes in special (Liquadol-PMT) developer.

The intensification characteristics shown are a function of the screen and film characteristics. In this connection, three factors are of prime significance in determining the shape of the curve in Figure 4. They are;

1. The absorption characteristics of the screens and air, within the wavelength range under consideration, are such that the absorption in the screens relative to that in air increases with decreasing wavelength.
2. The luminous efficiency of organic scintillation materials increases with increasing beta-ray energy (and hence photo- and recoil electron energy). According to Birks³⁵ the beta-ray excited fluorescence in organic crystals is due to exciton capture by single molecules, the excitons having been produced by the ionizing particle. Molecules damaged by the particles are believed to quench the excitons. In the low-energy range, where the specific energy loss of particles is high, the concentration of damaged molecules is correspondingly large and quenching is more pronounced than in the high-energy range.
3. The higher intrinsic sensitivity of photographic emulsions to light quanta than to charged particles further intensifies the two effects mentioned above. Conversion of the ionization energy in the screens to blue light (terphenyl emits between 3600 and 4300 Å) and subsequent highly efficient utilization of this energy by the film contributes greatly to the over-all intensification factor in the high-energy range.

A more detailed discussion of the fluorescence efficiency of the screens is given in Appendix 7.

The terphenyl screens were tested extensively in regard to stability by natural aging and by accelerated oven-aging. No changes were observed during either a period of 9 months storage at room temperature or after 14 days storage in a 60 C oven. A 1000-r exposure to radium gamma-rays also had no noticeable effect on the efficiency of the screens.

Before closing this section, it must be mentioned that the original molding experiments were conducted with anthracene, which yielded very efficient screens, when fresh. However, due to the presence of traces of the polymerization catalyst in the Lucite, the anthracene had a strong tendency to become oxidized and yellow with age and thus to lose its fluorescence efficiency.

C. Emulsion with Minimum Reciprocity Law Failure

The use of intensifying screens, while solving the wavelength-dependence problem, brought a new and different problem to the foreground. Whereas all commercial emulsions are known to have no reciprocity law

failure for x-ray exposures,* they usually exhibit considerable failure when exposed to visible light. This failure might amount to speed variations of a factor of 10 or more for the range of practical exposure times; in particular, the low-intensity failure is often very pronounced. The causes of reciprocity law failure have been the subject of much work because of its important implications in photographic theory; Webb and others³⁶ have studied many aspects in great detail.

Essentially, low-intensity reciprocity law failure, as demonstrated in the generalized curve of Figure 5, is due to the inefficiency of the process of latent image formation in its initial stage. According to Webb, a single latent image silver atom is not stable; for one particular emulsion he found a life-time of about 10 seconds. During a low-intensity exposure the first silver atom formed may lose an electron into the conduction band and the ion may migrate into an interstitial position before a second "stabilizing" silver atom is added to the sensitivity center. A high proportion of the absorbed energy can be lost in this manner; the latent image formation takes place only in a few highly effective sensitivity centers, that is, in the deepest traps in the crystal lattice. That this is actually the case has recently been demonstrated - in work not connected with this contract - by F. A. Hamm and the writer.³⁷ An electron microscopical study of grains which had received a slight physical development in a gold developer gave a very lucid demonstration of differences in latent image distribution and trap utilization for exposures to light of very low and optimum intensities.

On the basis of these observations one can conclude that for a minimum of low-intensity reciprocity law failure, an emulsion should consist of grains having a trap distribution in which deep traps are predominant. This requirement is in conflict with the requirements of low fog and good aging stability, because the deepest traps are believed to be those which fill up preferentially in the course of normal aging due to thermal agitation of the crystals. These would be the ones which subsequently initiate development without exposure.

The problem of designing an emulsion having a minimum of low-intensity reciprocity law failure was a difficult one. From the commercial point of view, very little attention had been paid to the effective response of emulsions in the 1 min to 40 hr exposure range. The literature contained, however, two valuable references. Trivelli and Loveland³⁸ had studied

*X-ray exposures of practically any duration are, in fact, high intensity exposures. The exposure of the individual grains takes place by photo- or recoil electrons (as discussed above) which travel at velocities of the order of magnitude of the velocity of light. The exposure time per grain is therefore extremely short, about 10^{-14} sec; the variation of exposure time as a function of electron energy and velocity is relatively small, and therefore one can consider all x-ray and gamma-ray exposures as one type of very high intensity exposure.

Fig. 5. Generalized Reciprocity Law Failure Curve.

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reciprocity law failure as a function of grain size and observed that reciprocity law failure decreased with increasing grain size. Jones and Hall³⁹ found in studies of effects of the duration of development that reciprocity law failure decreased with decreasing development time.

The proper approach to the development of an emulsion having minimum reciprocity law failure seemed to be the following:

1. To make an emulsion with predominantly large grains. Such an emulsion would probably have a tendency to yield undesirable high fog; however, it might be possible to counteract this by step 3.
2. To study sensitizing procedures which would lead to "deep" traps. Although a great deal is known in the photographic art about chemical sensitization methods, no precise information is available as to the trap depth produced by a particular method.
3. To provide sufficient speed for the purpose involved, in order to permit short or otherwise restrained development.

Since it appeared that wavelength independence could be more easily achieved by utilizing predominantly the light emitted by the screens, the need for low x-ray speed at all wavelengths was added as a further requirement. Therefore, it seemed advantageous to use an emulsion of the moderately fast, negative type. The following steps were taken in designing the emulsion to meet the requirements of the problem:

1. Selection of a slightly "foggy" gelatin.
2. Use of the highest tolerable making temperature, digestion time and after-ripening time.
3. Use of a high degree of gold sensitization.
4. Use of an indolizine-type stabilizer.

These measures resulted in an emulsion formulation which is described as M-Type (Monitoring Type) emulsion. It should be kept in mind, however, that such formulations should not be considered as strictly rigid. A certain amount of flexibility must be maintained, which depends a great deal on the selection of the gelatin. For example, the selection of a very "foggy" gelatin (rich in sensitizers and poor in restraining agents) may require a considerable downward adjustment in making temperature and second ripening time; possibly it may require also an increase in the amount of stabilization. On the other hand, the selection of a more inert gelatin may permit more extended heat treatment.

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The first major production trial was made without any stabilizing agent and consequently showed a fresh fog level of $D = 0.70$ for Liquadol development and $D = 0.30$ for restrained development. The reciprocity law failure was reduced from about 1000% for a commercial, Liquadol-developed emulsion to less than 100% for the M-Type emulsion with restrained development. However, the keeping quality of this emulsion was extremely poor.

In subsequent trials, less "foggy" gelatins were selected, and a moderate amount of stabilization was reintroduced. The resulting emulsion, represented by the M-Type formulation, had the reciprocity characteristics shown in Figure 6 and the keeping characteristics given in Table II.

TABLE II

Fog Stability of M-Type Emulsion
(Values for Trial P-81742)

Date	Age of film, days	Gross fog of film stored at 74 F in air condi- tioned area	Gross fog of film in contact with screen, with dura- tion of contact	Relative speed for white-light exposure
Film coated				
Nov 20, 1952	---	---	---	---
Jan 15, 1953	50	0.09	---	1.00
Jan 28, 1953	63	0.11	0.11 1 wk	1.05
Feb 4, 1953	70	0.11	0.11 2 wk	1.03
Feb 11, 1953	77	0.11	0.11 3 wk	1.08
Feb 18, 1953	84	0.12	0.12 4 wk	1.04
Mar 4, 1953	91	0.12	0.14 6 wk	0.98
Mar 18, 1953	105	0.13	0.14 8 wk	1.08
Apr 15, 1953	135	0.14	0.16 12 wk	0.98

Processed 3 minutes at 68 F in Liquadol containing 130 mg 1-phenyl 5-mercapto tetrazole per liter.

Fig. 6. Reciprocity law failure of M-type emulsion.

Curve A - Processed for 3 minutes in Liquadol

Gross fog
0.46

Curve B - Processed for 3 minutes in Liquadol + 130 mg/L of
1-phenyl 5-mercapto tetrazole

0.09

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Curve A shows the characteristics of the M-Type emulsion when processed in Liquadol for 3 min at 68 F (the fog is high in this case - about 0.45). Curve B shows the M-Type emulsion processed for 3 minutes at 68 F in Liquadol containing 130 mg of an anti-foggant, 1-phenyl 5-mercapto tetrazole,⁴¹ per liter. The magnitude of the improvement in reciprocity law failure is quite evident. The fresh fog (including base density) for this development is 0.09.

D. Development Restrainers

The term, "restrained development," used in the preceding paragraphs calls for some further elaboration. As mentioned before, a reduction in development time causes an appreciable loss in speed and a concurrent pronounced improvement in low-intensity reciprocity law failure. This is shown quantitatively for a typical emulsion in Figure 7. Development times of 1 min or less are, however, not very practical, and the use of various development restraining agents, so-called anti-foggants, was explored for this application. Several anti-foggants, such as benzotriazole and nitrobenzimidazole, are well known in the art; others are less familiar.

A large number of anti-foggants were studied in combination with M-Type emulsion; the results are presented in Table III. The efficiency of the compounds listed varies widely; in some cases very large quantities are needed to yield a useful effect. In choosing among these compounds the requirement of minimum reciprocity law failure should be considered first; low fog and satisfactory speed are second and third, respectively, in importance.

Another consideration is the availability of the compound; in this respect benzotriazole is advantageous. Although relatively large quantities are required, it produces a great reduction in reciprocity law failure, a slightly high fog, and very good speed. Higher concentrations of the anti-foggant or reduction of development time to 2 or 2-1/2 min, though not explored, may lead to a further improvement.

Of the compounds tested, tetrazolyl disulfide⁴⁰ had the best over-all characteristics - the fog is exceptionally low and reciprocity law failure and speed are acceptable. Unfortunately, this compound, when added to the developer, is slowly reduced to a mercapto tetrazole, thus causing a loss in reproducibility after standing approximately one-half hour. An aqueous solution of this compound can, however, be used with excellent results as a bathing treatment prior to development. The recommended use with M-Type emulsion and the results obtained are described in Table IV. The pre-bathing treatment has the advantage that the developer used for processing the monitoring film can be used later for processing ordinary x-ray film.

Fig. 7. Effect of development time on reciprocity law failure for a typical emulsion

				Gross Fog
Curve 1	-	1 minute	Liquidol processing time	.10
2	-	2	"	.11
3	-	3	"	.12
4	-	4	"	.14
6	-	6	"	.23

TABLE III

Characteristics of Anti-foggants When Used to Reduce
Reciprocity Law Failure in Monitoring Emulsions

Emulsion	Compound (added to Liquadol)	Concentration* mg/l	Reciprocity** law failure (%)	Gross Fog	Maximum speed (relative)
M-Type	None	-	350	0.46	100
M-Type	Nitrobenzimidazole	1100	170	0.08	28
M-Type	Thionalide	620	150	0.07	23
M-Type	5-allylamino 2-mercapto thiadiazole	200	150	0.13	33
M-Type	Mercaptobenzothiazole ⁽¹⁾	90	150	0.10	45
M-Type	Tetrazolyl disulfide ⁽²⁾	200	140	0.06	18
M-Type	4-ethyl 5-mercapto triazole	250	130	0.14	33
M-Type	Benzotriazole	3700	110	0.18	60
M-Type	1-phenyl 5-mercapto tetrazole	130	<100	0.09	32

1) Decreases D-max greatly

2) Unstable in developer; very useful as a pre-bath, see Table VIII

*mg/l Liquadol developer

**Per cent failure, i.e. speed differential between optimum intensity exposure (approximately 1 sec) and low intensity exposure (40 hr)

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TABLE IV

Pre-bath Treatment with Anti-foggants

1. Tetrazolyl disulfide

H ₂ O	1000 cc
Tetrazolyl disulfide	200 mg
Na ₂ CO ₃	30 g

Bathe for 3 min at 68 F before developing for 3 min in fresh Liquadol at 68 F.

2. 1-phenyl 5-mercapto tetrazole

H ₂ O	1000 cc
1-phenyl 5-mercapto tetrazole	200 mg
Na ₂ CO ₃	30 g

Bathe for 4 min at 68 F before developing for 3 min in fresh Liquadol at 68 F.

<u>Pre-bath</u>	Results obtained with M-Type emulsion		
	Reciprocity law failure (%)	Fog	Relative speed
None	350	0.46	100
Tetrazolyl disulfide	100	0.10	31
1-phenyl 5-mercapto tetrazole	100	0.13	32

As a developer addition, good results were obtained with 1-phenyl 5-mercapto tetrazole.⁴¹ It is stable and yields, for exposures to tungsten light, less than 100% reciprocity law failure, low fog and very satisfactory speed. This compound has been used in the final tests described in the next chapter. Its commercial use may require an analysis of the patent situation.

The mechanism of the interaction of the anti-foggants with the latent image and the causes for the reduction of reciprocity law failure have been the subject of considerable speculation in our laboratories but will be discussed elsewhere.

IV. RESPONSE OF THE FILM BADGE

A. General Construction

The double-coated M-Type film is placed between two dental-size screens of the composition described in Appendix 6. Since the screens emit light in both directions, the surfaces away from the film are to be covered with one sheet each of highly reflecting, glossy, baryta-coated paper stock (as used for the manufacture of photographic printing paper; BaSO_4 weight 0.007 mg/cm^2 ; paper weight 0.025 mg/cm^2). This sandwich is wrapped in black paper and inserted in the proper support. (It fits into the standard monitoring badge frame used by the Argonne National Laboratory, after removal of the metal filters.) This arrangement has the disadvantage of a rather complicated wrapping and unwrapping procedure.

To simplify this matter, the use of plastic cassettes (black nylon or other opaque plastic material) with permanently loaded screens is suggested. Samples of two types of cassettes were made in our workshop. One type uses dental-size screens and film; the other uses circular screens and circular film of larger diameter. Both constructions are shown in Figure 8. The cassettes are reasonably light-tight (negligible leakage during 1 hr exposure to direct sunlight); they are easy to load and unload and are very light in weight.

In the case of the dental-size cassette, it might be desirable to cover one part of the screens with black paper (see Figure 8), thus obtaining on the same piece of film areas exposed to both the direct and intensified radiation. This permits determination of the nature of the exposing radiation, if the exposure is great enough. No black paper is needed for the circular badge because of the larger diameter of the film relative to the screen.

B. Wavelength Dependence

The sandwich described in the preceding paragraph, wrapped in two layers of "Black Duplex Paper"* of 0.18 mm total thickness (equivalent to about 18 mg/cm^2), was tested in the range, $\lambda_{\text{eff}} = 0.60 \text{ A}$ to 0.11 A . The results for 1-hr exposures, made at the rate of approximately 0.10 r/hr , are shown in Figure 9 for the net $D = 0.4$. The film was processed for 3 min (with moderate agitation at 68F) in Liquadol containing 130 mg 1-phenyl 5-mercapto tetrazole** per liter of developer. The figure shows also the spectral sensitivity of the film, obtained by direct exposure (without intensification), and the intensification factor as a function of wavelength.

*Used in the photographic industry as rollfilm backing.

**Called PMT compound in future reference.

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Fig. 8. Exploded views of two types of monitoring badge cassettes showing complete contents of each; monitoring film is depicted as having been exposed and processed.

Dimensions of closed cassette:

Dental size cassette - 5.4 cm x 4.4 cm x 0.7 cm

Circular cassette - 4.3 cm diameter x 0.7 cm

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Fig. 9. Spectral sensitivity of M-Type emulsion (P-81742) with special Ansco Liquadol + PMT processing.

Speed measurements at net $D = 0.40$, for 1 hr exposures.

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The data are given also in Table V, which contains additional information on the radiation employed. The effective wavelengths were determined by the half-value layer method, using aluminum for the soft range and copper for the 250-Kvp radiation. In the case of radium gamma radiation the wavelengths assigned are 0.02 A for the unfiltered and 0.011 A for the filtered beam.

The speed values, in reciprocal roentgens, for the selected exposure rate and density level vary from a low of 7 r^{-1} to a high of 10 r^{-1} , or approximately $\pm 15\%$ from the midpoint. In reality, the variation might be larger, due to reciprocity law failure and slight changes in the shapes of the characteristic curves for exposures to radiation of different qualities.

TABLE V

Spectral sensitivities of M-type Monitoring Film P-81742; 1/r speeds for $D = 0.40$ above fog, exposed with p-terphenyl screens (6300 series); exposure time 1 hr. Processed in Liquadol + PMT, 3 min, 68F.

Tube	Kvp	Filtration	$\lambda_{\text{eff.}}$	1/r speed
Coolidge	65	Glass wall	0.60 A	7.0
Coolidge	100	Glass wall	0.52	7.6
OX-Industrial	65	Inherent*	0.39	7.8
OX-Industrial	150	Inherent*	0.30	9.8
OX-Industrial	150	1 mm Al	0.25	9.4
OX-Industrial	65	19 mm Al	0.24	7.6
OX-Industrial	150	2 mm Al	0.24	9.5
OX-Industrial	150	4 mm Al	0.23	9.1
OX-Industrial	100	0.13 mm Pb	0.21	9.2
OX-Industrial	250	Inherent*	0.21	9.9
OX-Industrial	150	10 mm Al	0.20	8.9
OX-Industrial	150	21 mm Al	0.16	9.0
OX-Industrial	250	21 mm Al	0.13	9.8
OX-Industrial	250	2.5 mm Cu	0.10	9.6
OX-Industrial	250	"Standard"**	0.07	9.2
OX-Industrial	250	"Heavy"***	0.06	8.9
Ra	Gamma	None	0.02	9.6
Ra	Gamma	2.0 cm Pb	0.011	9.8

*The inherent filtration of this equipment is equivalent to 3 mm Al at 100 Kvp.

**A composite filter of 0.5 mm Pb, 4.1 mm Cu, and 4.8 mm Fe

***A composite filter of 1.5 mm Pb, 8.2 mm Cu, and 1.6 mm Fe

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C. Reciprocity Law Failure

As discussed earlier, the variation in speed due to reciprocity failure in the 1-sec to 40-hr exposure range amounts to about 100% for M-Type emulsion processed in Liquadol containing 130 mg PMT per liter. The routine testing for reciprocity law failure which led to this result was done with white light. In the actual use of the film with intensifying screens, the exposure is a mixture of x-radiation, gamma-radiation, and light. However, the light is emitted in very intense pulses, each pulse corresponding to the absorption of an x-ray or gamma-ray quantum by the screens. This is expected to decrease the low-intensity failure to a certain, unknown extent. The effect should be most pronounced in the gamma-ray range, for evident reasons. The M-Type emulsion, in combination with the intensifying screens was therefore tested for reciprocity law failure in actual gamma-ray exposures over the range of 15-sec to 40-hr exposure duration, the corresponding exposure rates for the $D = 0.40$ level varying from about 25 r/hr to 2 mr/hr. As shown in Figure 10, the reciprocity law failure decreased noticeably from 100% for exposure to white light, to 65% for the exposure to gamma-rays with screens. The characteristic curves for gamma-ray exposures corresponding to the extremes of the reciprocity curve are shown in Figure 11. For radiation of longer wavelength the effects of reciprocity failure must be less than this amount, because of the increasing contribution of the direct x-radiation.

D. Characteristic Curves - Their Shapes

It is known that the shapes of the characteristic curves of photographic emulsions are different for exposures to light and x-rays. Exposures to gamma-radiation yield curves similar to the x-ray curves but deviating slightly in the direction towards the steeper characteristic obtained with light. Generally, x-ray exposures produce a flatter toe and a higher D-max in comparison with those obtained with a light exposure. The differences are minimized in emulsions of large average grain size and narrow grain-size distribution. The need for such characteristics was recognized early. Fortunately they were in line with the requirement of minimum reciprocity law failure. The M-Type emulsion exhibits, therefore, a minimum difference between the characteristic curves for x-ray and light exposures.

E. Extremes of Sensitivity

It seemed desirable to learn the conditions under which the badge would exhibit its lowest and its highest sensitivity. The extremes in the low density range ($D = 0.4$) are:

Minimum sensitivity: $\lambda_{\text{eff}} = 0.60 \text{ A}$, 65 Kvp Coolidge Tube, glass wall, exposure time 40 hr, exposure rate 0.004 r/hr

Maximum sensitivity: $\lambda_{\text{eff}} = 0.21 \text{ A}$, (250 Kvp unfiltered), exposure time 5 min, exposure rate 1.2 r/hr

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Fig. 10. Comparison of reciprocity characteristics of M-type emulsion (P817/42) as determined by radium gamma-ray terphenyl-intensified exposures and normal white-light exposures.

Fig. 11. Characteristic curves of M-type emulsion (P81742) for radium gamma-ray exposures.
Source - 198.6 mg. radium in Al. capsule, effective wavelength = 0.02 Å.

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The extremes in the high density range $D = 1.8$ are:

Minimum sensitivity: $\lambda_{\text{eff}} = 0.02 \text{ A (Ra-gamma)}$, exposure time
40 hr, exposure rate 0.09 r/hr

Maximum sensitivity: $\lambda_{\text{eff}} = 0.21 \text{ A (250 Kvp unfiltered)}$, exposure
time 5 min, exposure rate 18 r/hr

The characteristic curves corresponding to the above data are plotted in Figure 12.

For exposure rates higher than those indicated for "maximum sensitivity," the sensitivity will be less because of high-intensity reciprocity law failure. This type of failure is usually less than the failure in the low-intensity region; therefore, one does not expect the sensitivity to decrease below the "minimum sensitivity" values given. Actual tests of the high-intensity failure could not be conducted because of the lack of a suitable high-intensity gamma-ray or x-ray source.

F. Normalized Curve

It is not intended to give more than general recommendations for the actual use of the badge. The laboratories will have to adapt their calibration and evaluation procedure to their own needs. It is recommended to use a calibration curve corresponding closely to the type of radiation to which the user is most likely to be exposed. This will reduce the probable error of evaluation to a minimum. If no preference exists, an average curve halfway between the extremes can be taken as a "normalized" curve. In doing this we arrived at the idealized curve shown in Figure 13. Using this curve one would arrive at evaluations of the following sort:

Net density	Evaluated dose	Possible extremes of true dose	Possible error
$D = 0.40$	135 mr	180 mr 92 mr	$\pm 32 \%$
$D = 1.0$	480 mr	610 mr 340 mr	$\pm 29 \%$
$D = 1.5$	1220 mr	1600 mr 820 mr	$\pm 33 \%$

The maximum interpretation error under these conditions is of the order of magnitude of $\pm 30 \%$. The errors which are inherent to the photographic process (processing, densitometry, etc.) are only partially included in this value. Their determination would require an extensive statistical

Fig. 12. Characteristic curves of M-type emulsion (P81742) for extreme values of exposure rate and sensitivity.

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Fig. 13. Suggested normalized characteristic curve for monitoring film P81742.

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evaluation which has not been conducted. The magnitude of such errors depends of course on the quality of the processing and on the control methods employed.*

G. Possible Improvements and Modifications

The data supplied in this general chapter are those obtained with the experimental M-Type emulsion coating P-81742. One minor weakness of this coating is a slightly low D-max. This can be corrected in future coatings and in production by increasing the coating thickness by 1 to 2 microns on each side, thus permitting improved interpretation in the 5-10 roentgen range.

The reciprocity characteristics can also be improved by omission of the stabilizer from the M-Type formulation. This will adversely affect the keeping quality of the emulsion and may cause a fog increase of as much as 0.04 density unit per month for storage in an air-conditioned room. However, if desired, this emulsion deterioration with age can be drastically reduced (to $\Delta D = 0.01$ per month, or less) by storage in an ordinary household refrigerator.

* Another source of error which would be encountered in practice is the directional response of the badge. This aspect is discussed further in Appendix 8.

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V. PATENT ACTION

In compliance with Article VI - Patents under this contract, the Atomic Energy Commission was notified by the Patent Counsel of the Ansco Division in a letter dated May 5, 1953 to determine whether patent applications should be filed on the following three proposals:

- AD# -1196 - Organic Fluorescent Intensifying Screens Having High Response to Gamma Radiation - Herman Hoerlin and Frank J. Kaszuba (Argonne-Ansco #1)
- AD# -1197 - Fluorescent Intensifying Screens Containing Terphenyl - Frank J. Kaszuba and Herman Hoerlin (Argonne-Ansco #2)
- AD# -1198 - Monitoring Film Badge Having Wavelength Independent Response in the 2 mev to 40 kv Range - Herman Hoerlin, Frank J. Kaszuba, Robert H. Clark, Ernest T. Larson, Jr. (Argonne-Ansco #3)

A copy of this letter is given in Appendix 9.

The decision of the Atomic Energy Commission as to the disposal of these patent proposals should be forwarded to Mr. E. P. Gilheany, Division Patent Counsel, Ansco, Binghamton, N. Y.

ACKNOWLEDGMENTS

In addition to those listed as staff members, thanks are due to Dr. F. W. H. Mueller for many helpful suggestions, to Mr. Stanley Popeck for contributions to the solution of the problem of screen manufacture, to Dr. Victor Hicks for assistance in the formulation of the project, to Mr. James F. Ruddy, Jr. for assistance in the studies of reciprocity law failure, and to Mr. E. T. Larson for his assistance in editing the final report.

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APPENDIX 1

Procedure for Surface Development

1. Develop 4 min at 68F in the following:

Na_2CO_3	120 g
Water, distilled and boiled, to make	1 liter
Glycin, added just prior to development	24 g

2. Short stop, fix and wash as normally.

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APPENDIX 2

Procedure for Internal Development

1. Wash 15 min in running water.
2. Bleach 10 min in 0.25% CrO_3 at 68F.
3. Wash 15 min in running water.
4. Develop 5 min at 68F in the following:

A-125 Developer	1 liter
Sodium thiosulfate	8 g
Potassium iodide	0.1 g
Sodium bromide (only if needed to inhibit fog)	4 g

5. Short stop, fix and wash as normally.

Formulation for A-125 Developer:

Hot water (125F)	750 cc
Metol	3 g
Sodium sulfite, anhydrous	44 g
Hydroquinone	12 g
Sodium carbonate, monohydrate	65 g
Potassium bromide	2 g
Water, to make	1 liter

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APPENDIX 3

Electron Sensitization with Organic Compounds

The following compounds showed moderate positive results:

15% increase in gamma-ray and 65-Kvp x-ray speed.

10% increase in gamma-ray speed.

5% increase in gamma-ray speed,
15% improvement in 65-Kvp/gamma-ray speed ratio.

65-Kvp/gamma-ray speed ratio improved by 5%.

65-Kvp/gamma-ray speed ratio improved by 10%.

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APPENDIX 4

Small Batch Recrystallization Purification Process for Terphenyl

Approximately 400 grams of terphenyl* are dissolved in 2.5 liters of redistilled toluene. This solution is placed in a 5-liter flask having a reflux condenser attached and heated for at least 15 minutes. The hot solution is rapidly filtered; the filtered solution is then cooled quickly in an ice water bath. During this cooling, and with constant stirring, small terphenyl crystals precipitate from the solution. These precipitated crystals are removed by filtration, redissolved in more redistilled toluene, then heated again, etc., as described above. This procedure is repeated until the third cycle of recrystallization is completed. Further recrystallization of the terphenyl is not warranted.

*1,4-diphenylbenzene (purest grade), M.P. 212-213 C, listed as item #2103 in organic chemicals catalog of Eastman Kodak Company, Rochester, New York

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APPENDIX 5

Manufacture of Terphenyl-Lucite Intensifying Screens

In general, the laboratory method of manufacturing the intensifying screens consists of the compression molding of a mixture containing the scintillation compound terphenyl dispersed in powdered Lucite.

For preparation of the mixture for molding, the 3-cycle recrystallized terphenyl crystals were pressed through a 40-mesh stainless steel screen to insure against large crystals or clusters of crystals. (The more uniform the particle sizes, the more uniform the resultant screen and consequently the more uniform the intensified exposure of the monitoring film.) The Lucite powder,* as obtained, had a large size distribution and was therefore sifted over a 40-mesh stainless steel screen; all Lucite particles not passed by this screen were discarded. This sifted Lucite powder was subsequently resifted over an 80-mesh stainless steel screen; the Lucite particles passing through this screen were used for screen laminations (described later). The Lucite particles not passed by the 80-mesh screen were used in the terphenyl-Lucite mixture.

For the final mixture, in the ratio of 2 parts terphenyl to 1 part Lucite, by weight, terphenyl was added to the Lucite, these compounds being mixed together thoroughly. Inasmuch as Lucite and terphenyl have a high tendency to become electrostatically charged, the final mixture was poured into a bottle, which was filled about three-quarters full, and subsequently tumbled in an high-intensity 250-Kvp x-ray beam for approximately 1 hr. The ionization produced by the absorption of the x-rays in the mixture caused a neutralization of the electrical charges on the particles and permitted a much more uniform mixture.

The mold used for the compression molding of the screens was made from #316 stainless steel, with the surfaces that determined the molded screen's contours being highly polished. Compression molding requires both pressure and heat to convert the terphenyl-Lucite powder mass into a solid object - the intensifying screen. Holes were drilled in the upper and lower sections of the mold to provide for the insertion of cartridge-type heaters. An additional hole was drilled in the lower section to provide for the insertion of a stem thermometer. Because of its proximity to the heaters, this thermometer does not necessarily indicate the true temperature of the mold's contents but does provide a very effective means of controlling the temperature. Pressure was applied to the mold by means of a laboratory hydraulic press, the two platens of the press having first been carefully leveled to provide for uniform thickness throughout the molded screen.

*Lucite Acrylic Resin Moulding Powder, procured from E.I. duPont de Nemours and Co., listed as item #H7500 HG29, medium-flow colorless transparent granular polymer.

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As previously mentioned, the intensifying screens were laminated with Lucite, that is, a thin layer of Lucite was molded to each side of the intensifying screen. This lamination was used for several reasons, mainly to provide added strength to the screen so that it could better withstand physical abuse without damage. The lamination of a 0.005 in. layer of Lucite to each side of a screen increased the resistance to breakage by a factor of 5. (Because of the comparatively low amount of Lucite carrier used in the terphenyl-Lucite mixture, the screens made without laminations were too fragile for general use.) These laminations also provided a tough surface over the screens, making them much less susceptible to surface scratches. Also, the laminations increased slightly the luminous efficiency of the screen and, in addition, resulted in a much more uniform density on the exposed film.

The procedure for loading the mold and the actual molding of the screens is as follows:

1. Pour 200 mg of Lucite powder (passed by an 80-mesh screen) into the mold and level to a thin, smooth layer. (This amount of Lucite will form a 0.005 in. lamination layer.*)
2. Pour 2.7 g of the terphenyl-Lucite mixture into the mold over the level layer of Lucite.
3. Level the mixture, taking care not to disturb the Lucite layer underneath.
4. Compress these two layers by means of hand pressure, using a plastic or metal tamper which is slightly smaller than the mold cavity.
5. Repeat step (1) for the addition of the final Lucite lamination layer.
6. Close the mold and place it between the platens of the hydraulic press, applying only enough pressure to hold the mold in place.
7. Turn on the heating cartridges. When the thermometer indicates a temperature of 120C, apply a pressure of 530 lb/sq in. to the mold.
8. While the mold is under this pressure, allow the temperature to continue to increase. Increase the pressure to 2,650 lb/sq in. when the temperature reaches 130C.
9. Maintain this pressure until the temperature reaches 160C. The heating units should then be turned off and the mold cooled by air from a large-volume fan. During the cooling, the pressure applied to the mold should no longer be adjusted; it will decrease slightly as the mold cools.
10. After cooling to approximately 45C, the mold can be removed from the press and opened for the removal of the screen.

*The quantities described are for the formation of a dental film size intensifying screen.

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APPENDIX 6Physical Characteristics of
Terphenyl-Lucite Intensifying Screen

Dimensions:

Length	4.07 cm
Width	3.09 cm
Thickness	0.22 cm
Hence, area	12.24 cm ² (1/4 in. r corners)

Composition:

Total weight of screen	3.1 g
Total weight of Lucite lamination (both sides)	0.4 g
Total weight of terphenyl-Lucite mixture	2.7 g

In terphenyl-Lucite mixture,

Total weight of terphenyl	1.8 g
Total weight of Lucite	0.9 g

Mass per unit area:

Screen	253 mg/cm ²
Terphenyl	147 mg/cm ²
Lucite	106 mg/cm ²

Transmission:

The spectrophotometric transmission density curve of a single screen is shown in Figure 14.

Fig. 14. Transmission density curve for type 630C terphenyl screen. This curve was determined with the sample in immediate contact with the integrating sphere of a General Electric recording spectrophotometer so that essentially all of the transmitted light is collected and measured.

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APPENDIX 7Calculation of the Fluorescence Efficiency of the
Terphenyl-Lucite Screens

The following calculation of fluorescence efficiency is based on a theoretical calculation of the energy truly absorbed by the screens per unit exposure in roentgens and the experimentally determined values of the exposure in roentgens needed to produce a density of 0.4 on the monitoring film when exposed with and without screens.

The calculation of the energy truly absorbed per incident roentgen is based on the following considerations. From the definition of the roentgen,

$$E_0 = \frac{85 \text{ r}}{1 - e^{-\left(\frac{\mu}{\rho} - \frac{\sigma_s}{\rho}\right)}}$$

where

E_0 = incident energy in ergs/cm²

r = number of incident roentgens

μ/ρ = mass absorption coefficient for air

σ = total scattering coefficient

σ_a = recoil coefficient

$\sigma_s = \sigma - \sigma_a$ = coefficient of true scattering for air

$\frac{\mu}{\rho} - \frac{\sigma_s}{\rho}$ = coefficient of true mass absorption for air

For the screens, the true absorbed* energy is given by:

$$E_{\text{abs}} = E_0(1 - e^{-\mu'x}) \left(1 - \frac{\sigma'_s}{\mu'}\right)$$

E_{abs} = energy truly absorbed

x = total thickness of screens

μ' = mass absorption coefficient for the screens

σ'_s = coefficient of true scattering for the screens

The values of μ/ρ for air and the various elements composing the screens were taken from the published data of Victoreen.⁴² The over-all value of μ'/ρ' for the screens was computed by combining the values of μ'/ρ' for the constituent elements, each weighted according to its contribution to the total mass of the screens

*Neglecting the slight absorption by the film between the screens.

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The calculation of the values of σ_s and σ_s' is more involved:

$$\sigma/\rho = \sigma(e) N_o \frac{Z}{A}$$

where,

$\sigma(e)$ = total scattering coefficient per electron

N_o = Avogadro's number

Z = atomic number of the material

A = atomic weight of the material

A chart of values for $\sigma(e) N_o$ as a function of wavelength is also given by Victoreen. The value of $\sigma(e)$ is related to the classical value of the scattering by a single electron by the Compton cross-section :⁴³

$$\sigma(e) = \frac{\sigma_o}{1 + 2\alpha}$$

where,

σ_o = classical scattering by a single electron, and

$$\alpha = \frac{h\nu}{mc^2}$$

(This expression is not very accurate for $\frac{h\nu}{mc^2} > 1$, but is probably sufficiently accurate for the present purpose.)

$\sigma(e)$ is composed of two parts:

$$\sigma_a(e) = \frac{\sigma_o}{(1 + 2\alpha)^2}, \text{ and}$$

$$\sigma_s(e) = \frac{\sigma_o (1 + \alpha)}{(1 + 2\alpha)^2}$$

Hence,

$$\frac{\sigma_s(e)}{\sigma(e)} = \frac{1 + \alpha}{1 + 2\alpha},$$

or

$$\frac{\sigma_s}{\rho} = \sigma(e) N_o \frac{Z}{A} \frac{(1 + \alpha)}{(1 + 2\alpha)}$$

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Values of $\frac{\sigma_s}{\rho}$ for air and for the screen were calculated for a series of wavelengths, followed by an evaluation of E_0/r and E_{abs}/E_0 . The product of the latter values then gave E_{abs}/r , or the truly absorbed energy, in ergs/cm², per incident roentgen at each wavelength.

These results were then combined with the experimental data on the exposure required to produce a net density of 0.4 for intensified and direct (screens present but covered with black paper) exposures. If, for the direct exposure, r_1 roentgens are required to yield a net density of 0.4, and if r_2 roentgens are required to produce the same density in the intensified exposure, then the amount of light emitted by the screens per incident roentgen is proportional to $\frac{r_1 - r_2}{r_1 r_2}$. This expression is based on the assumption that the exposure due to the fluorescent light from the screens and the exposure due to direct x-irradiation of the film follow the van Kreveld⁴⁴) law of additivity for all x-ray wavelengths. The relative amount of light emitted by the screens per unit of energy truly absorbed by the screens is then given by:

$$\frac{\frac{r_1 - r_2}{r_1 r_2}}{r_2 \frac{E_{abs}}{r}} = \frac{\frac{1}{r_2} - \frac{1}{r_1}}{r_2 \frac{E_{abs}}{r}}$$

The expression $\left(\frac{1}{r_2} - \frac{1}{r_1}\right)$ is just the difference in speed, expressed in reciprocal roentgens, between intensified and direct exposures.

Table VI gives the results at several stages in the calculations. The results given are for a pair of the standard screens, each containing 147 mg/cm² of terphenyl and 107 mg/cm² of Lucite. Figure 15 gives the curves for the truly absorbed energy per incident roentgen and the relative fluorescence per unit of truly absorbed energy.*

*These results are based, in part, on calculations made by Ruth Seiwatz.

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TABLE VI

Stages in the Calculation of Fluorescence Efficiency

Wave-length A	$\frac{\mu}{\rho}$ (air)	$\frac{\mu'}{\rho'}$ (screens)	$\frac{\sigma_s}{\rho}$ (air)	$\frac{\sigma'_s}{\rho'}$ (screens)	$\frac{E_o}{r}$, ergs/cm ² per r	$\frac{E_{abs}}{E_o}$ (screens)	$\frac{E_{abs}}{r}$, ergs/cm ² per r (screens)	$\frac{1}{r_2} - \frac{1}{r_1}$, roentgens ⁻¹ (film & screens)	$\frac{1}{r_2}$, roentgens ⁻¹ (film & screens)	Relative fluorescence efficiency, arbitrary units (screens)
.02	.0793	.0849	.0515	.0550	3060	.0144	44.2	10.4	10.6	2.50
.04	.1052	.112	.0760	.0812	2820	.0147	41.5	9.2	9.2	2.04
.08	.1337	.142	.1075	.115	3120	.0133	41.5	7.2	8.4	1.46
.10	.1433	.152	.1180	.126	3360	.0127	42.6	6.3	8.2	1.21
.20	.1862	.185	.1465	.158	2140	.0129	27.6	3.5	8.2	1.04
.40	.3561	.283	.1710	.182	509	.0476	24.2	2.55	7.9	.83
.60	.7750	.500	.1795	.192	190	.137	25.9	1.43	5.7	.314

Fig. 15. Relative efficiency of fluorescence and energy truly absorbed per incident roentgen for a pair of the terphenyl-Lucite intensifying screens.

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APPENDIX 8Directional Effect of Radiation on the Response
of the Monitoring Badge

Monitoring badges* were exposed to 65-Kvp x-rays and to radium gamma-rays, the individual badges being arranged so that the angle between the plane of the badge and the source-badge axis was respectively 90, 60, 30, and 0 degrees, as indicated in Figure 16. The source-badge distances were sufficiently large so that the effect of distance variation between the source and various points on the badge would be negligible.

For the evaluation of the exposed, processed film, the central portion of the intensified-exposure area was used. Much less deviation in response could be obtained by using the "leading-edge" or the highest density area of the exposed film.

The directional effect of radiation on the response of the monitoring badge is shown graphically in Figure 16. The effect is very slight for hard radiation because of the comparatively low absorption of the intensifying screen for this type of radiation. Were this a metallic-filter type of monitoring badge, it is obvious that the directional effect would be quite drastic. As expected, the directional effect was more pronounced for 65-Kvp x-rays because of the relatively higher absorption of the intensifying screen for this type of radiation. A maximum decrease in sensitivity of 37% was caused by the directional effect when the badge was exposed to the soft radiation.

* These badges were wrapped in black paper instead of being housed in the Nylon plastic monitoring badge cassettes, inasmuch as these cassettes were devised after this experiment was conducted.

Fig. 16. Directional effect of radiation on the response of the monitoring badge.

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APPENDIX 9

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Patent Correspondence

May 5, 1953

Foster York, Chief
Chicago Patent Group
U.S. Atomic Energy Commission
P. O. Box 299
Lemont, Illinois

SUBJECT: ANSCO SUBCONTRACT W-31-109-ENG-38 WITH ARGONNE
NATIONAL LABORATORY

Dear Sir:

Please refer to your letter of April 23, 1953, to our Dr. Hoerlin on the above subject.

In pursuance of Article VI, paragraph 1 of the above identified Agreement, I am annexing the Ozalid masters of three (3) patent disclosures, which are identified as:

Argonne-Ansco No. 1 (AD-1196)

Argonne-Ansco No. 2 (AD-1197)

Argonne-Ansco No. 3 (AD-1198)

It would appear that these disclosures are so related that they could be combined to form a single patent application.

You will note that much of the work covered by these disclosures has been reported in the prior art. It is, therefore, problematical if a strong patent can be obtained on the development in question.

Nevertheless, should you decide to file applications on the disclosures, such applications should be filed in the names of the following parties as indicated for each disclosure:

Argonne-Ansco No. 1 - Herman Hoerlin, c/o Los Alamos Scientific Laboratory, University of California, Post Office Box 1663, Los Alamos, New Mexico; and Frank J. Kaszuba, 7 Matthews Street, Binghamton, New York;

To: Foster York

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-2-
5/15/53

Argonne-Ansco No. 2 - Frank J. Kaszuba, 7 Matthews Street,
Binghamton, New York; and Herman Hoerlin, c/o Los Alamos
Scientific Laboratory, University of California, Post Office
Box 1663, Los Alamos, New Mexico;

Argonne-Ansco No. 3 - Herman Hoerlin, c/o Los Alamos Scientific
Laboratory, University of California, Post Office Box 1663,
Los Alamos, New Mexico; Frank J. Kaszuba, 7 Matthews Street,
Binghamton, New York; Robert H. Clark, 14 Highland Road,
Chenango Bridge, New York; and Ernest T. Larson, Jr., 3 Sears
Avenue, Binghamton, New York.

If I can be of further assistance, kindly advise.

Very truly yours

E. P. Gilheany
Division Patent Counsel

EPG:ij

Enclosures

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